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16. ABSTRACT

The Clean Air Act Amendments of 1970 authorized the U.S. Environmental Protection Agency (EPA) to conduct research on the effects of air pollutants on the service life of materials. Information from the materials effects research program will be used by the Agency to conduct cost-benefit studies and to establish criteria for developing secondary air quality standards. This report provides an overview of the materials effects research program that EPA conducts for the National Acid Precipitation Assessment Program (NAPAP) and the testing facilities and technical procedures that are being developed for characterizing environmental degradation of materials, especially coatings. As such it is in the national interest to share this knowledge and these new capabilities with industry. Because the NAPAP research is to be completed by 1990, the testing facilities at EPA will be available for use and funding support by organizations outside the Agency. In 1986, Congress passed the Federal Technology Act, Public Law No. 99-502, that permits EPA to enter into Cooperative Research and Development Agreements with private industry and other organizations, thereby utilizing their facilities for environmental testing and development of materials.

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Research News

Review of EPA Materials Program: Environmental Testing

By J.W. Spence, E.O. Edney, and F.H. Haynie,
US Environmental Protection Agency, Atmospheric Research and Exposure Assessment Laboratory

The Clean Air Amendments of 1970 authorized the US Environmental Protection Agency (EPA) to conduct research on how air pollution affects the service life of materials. Information from this research will be used for cost-benefit studies and as criteria for developing secondary air quality standards by the EPA.

This report provides an overview of the research program that EPA conducts for the National Acid Precipitation Assessment Program (NAPAP) and the testing facilities and technical procedures that are being developed for characterizing environmental degradation of materials, especially coatings.

NAPAP Materials Effects Program

The Acid Precipitation Act of 1980 established an interagency task force to plan and implement a 10-year national program on acidic deposition research. This task force implemented NAPAP by forming Task Groups to investigate the various aspects of acidic deposition. Task Group VII, Effects on Materials and Cultural Resources, was formed to study the effects of acidic deposition on man-made and cultural materials. A listing of the materials that the Task Group is studying for potential damage due to acidic deposition is shown in Table 1. The federal agencies that are responsible for planning and conducting effects research on these materials are also shown. A general description of the exposure program and testing procedures has been previously reported.[1] The primary mission of Task Group VII is to provide NAPAP with dose-response relationships for the incremental effects of acidic deposition on the materials at risk. This information, along with estimates of the geographic distribution of materials, etc., will be incorporated into State of Science and Materials Assessment Reports that are due to Congress at the completion of the NAPAP in 1990.

Coatings Research Program

As shown in Table 1, the EPA is responsible for studying the effects of acidic deposition on coatings within the NAPAP. In previous EPA exposure studies of coatings, an erosion technique was used to investigate the effects of pollutants on house and maintenance coatings.[2,3,4] This technique, in

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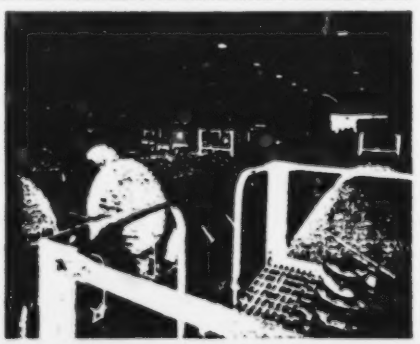

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Table 1 Materials Exposed by Task Group VII for NAPAP

Materials	Agency
Metals 1010 Carbon Steel Cor-Ten A Steel 110 Copper 3003 H14 Aluminum 191 Zinc G-90 Galvanized Steel Galvalume	Bureau of Mines
Exterior Paints Coatings on metal Coatings on wood/products	US Environmental Protection Agency
Dimension Stone Salem limestone Vermont marble	National Park Service and US Geological Service

which the coating is applied to an inert substrate, does not permit an investigation of film failures involving various substrates. For the NAPAP study, EPA prepared a research plan for investigating the effects of acidic deposition on film-substrate systems.[5]

An important part of this plan involved the preparation of a report on the types and application of exterior paint used in the United States and the failure modes of economically important coating substrate systems.[6]

The report recommends that acrylic latex on wood as well as alkyd on steel be investigated to determine whether acidic deposition has a role on the observed failure modes of these coated systems. Coated steel was included in this study because the steel substrate is susceptible to accelerated corrosion by pollutants, and undercutting or rusting is known to promote repainting.

Based on information in this report and the recommendations of experts at an EPA coatings workshop that was held in 1985[7], the following coating-substrate systems were selected to initiate the exposure study:

- alkyd coating on hot rolled carbon steel,
- acrylic latex coatings on western redcedar, and
- alkyd coating on western redcedar.

Exposure of these coated substrates initially involves 2 phases of research:

- characterization of atmospheric deposition to coating surfaces and the reaction products, and
- development of state-of-the-art analytical techniques to determine whether acidic deposition promotes film-substrate damage.

Pending the results of these investigations, exposure experiments will be designed from which preliminary dose-response relationships may be developed.

To characterize deposition and the interactions of atmospheric pollutants with coatings as well as other materials, NSI Environmental Sciences under contract to EPA developed unique chamber and field exposure facilities.

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Environmental Chamber

For this study, an environmental exposure system was developed with the capability of exposing materials to light/dark-condensation cycle in the presence of simulated urban environments and other complex air mixtures. This chamber has some characteristics of an earlier chamber developed by EPA.[8] A diagram of the exposure system that consists of 2 material exposure chambers coupled to a 11.3 m^3 chamber for generating photochemical smog is shown in Fig. 1. The system operates as a continuously stirred tank reactor with the large chamber serving as a reservoir for the exposure chambers.

The composition of the photochemical smog mixture is continuously monitored and is maintained at a desired level by adjusting the flow rate of the reactants and the residence time of the smog chamber. Materials within the flow exposure chambers are exposed to photochemical smog of known chemical composition. The 2 flow exposure chambers are connected with glass piping to the photochemical chamber. Blowers are used to maintain turbulent conditions at wind speeds of approximately 3.0 meters/second (m/s) in each chamber. A cross sectional drawing of an exposure flow chamber is shown in Fig. 2.

The exposure chambers are equipped with lamps to simulate sunlight during the exposure cycle. Both exposure chambers have 7 positions for mounting test panels. Test panels are mounted to a chiller back plate that permits the temperature of the panels to be lowered below the dew point temperature of the chamber, thus forming condensation on the test panels.

Collection and analysis by ion chromatography and atomic absorption spectroscopy of the condensation permit the identification of the gaseous species that deposit within the smog mixture and the soluble products that form through reactions with the test panel. Within a 24-hour exposure period, considerable insight is acquired concerning the deposition of gases to material test panels in the presence and absence of surface moisture.

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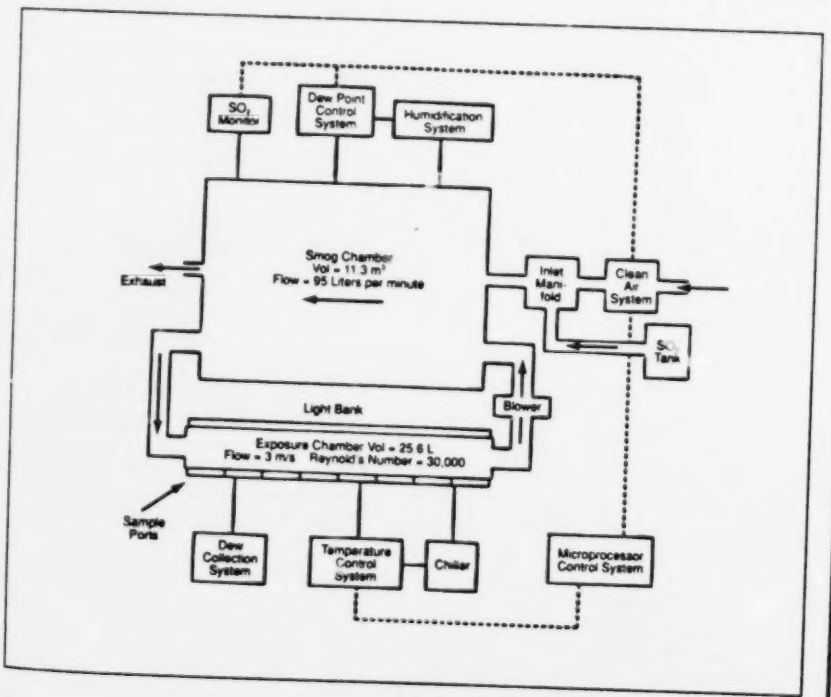

■ Fig. 1
EPA Environmental Materials Exposure System

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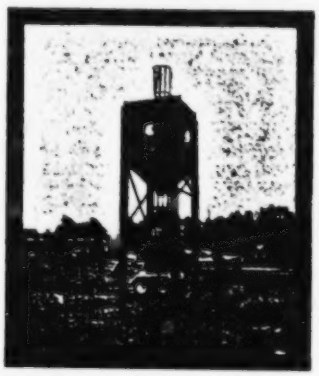

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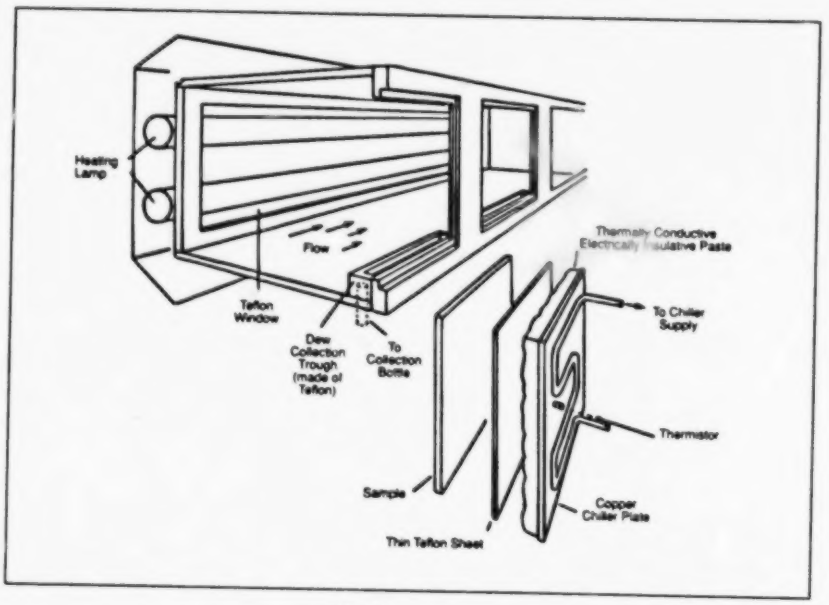
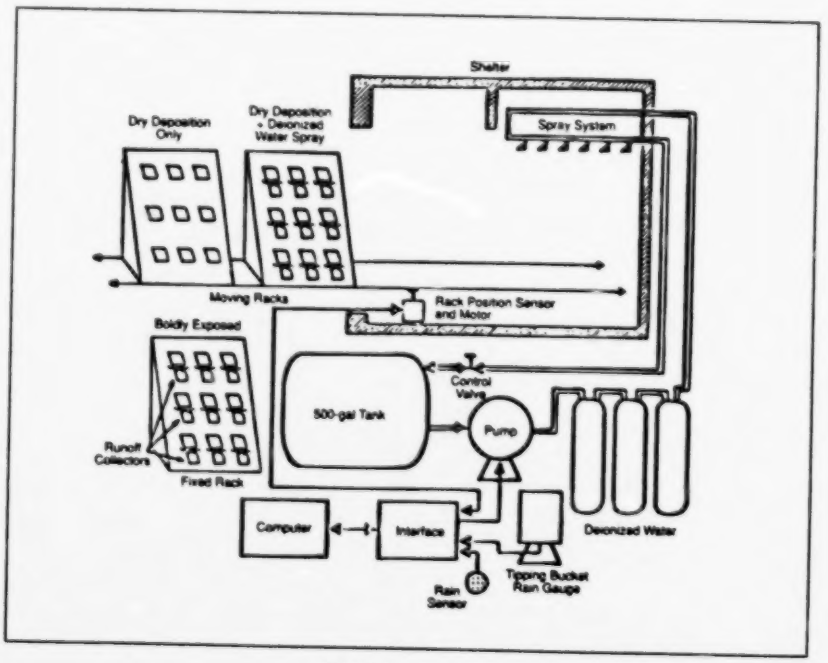


■ Fig. 2
Cross-sectional diagram of Materials Exposure Flow Chamber



■ Fig. 3
Automatic covering-spray device

A second chamber, designed and constructed by NSI—Environmental Sciences for EPA, was recently installed at North Carolina State University. This chamber has over 3 times the capacity for exposing materials than the system now being used. Long-term exposures of the coating-substrate systems for deriving preliminary dose-response relationships are planned with this chamber.

Field Exposures

When Task Group VII began the Materials Effects Research Program, it established 5 field sites located in the eastern, northeastern, and midwestern US for boldly exposing the materials shown in Table 1.

In a prior EPA materials field exposure study, statistical regression techniques showed that the effects of pollutants on coating were masked by strong covariances with other environmental parameters.[4] The range of rain acidity (pH) at the 5 sites was not large.

In order to enhance the study of the effects of wet and dry acidic deposition on boldly exposed materials, automatic covering devices that not only cover a set of test panels but also spray an identical set with deionized water of pH 5.6 during each rain event were installed at the Research Triangle Park, NC and Steubenville, OH exposure sites. After each rain event, the moveable racks are automatically returned to the boldly exposed position. The use of this device, as diagrammed in Fig. 3, has proven to be an effective means of separating the effects of wet and dry acidic deposition on materials.[9]

The deionized spray of these devices delivers approximately the same volume of water that falls for each rain event. However, the spray does not simulate rain intensity or direction of rainfall. Collection bottles are mounted on the exposed material panels to collect rain and spray runoff for each rain event. The runoff collections are analyzed in the same way as the condensation samples that are obtained from panels exposed in the environmental flow chamber system. The analysis of the spray collections provides information on the effects of the dry deposition process, i.e., reaction of gases during periods of surface wetness (dew) and dryness. The analysis of rain runoff collections from the boldly exposed test panels provides information on the effects of both the dry and wet (rain acidity) deposition. Since the runoffs are analyzed for each rain event, information on the effects of atmospheric deposition on the test panels is acquired in the earlier stages of field exposure.

Characterizing Acid Deposition

Major findings concerning the characterization of acidic deposition to coating and other materials that have been obtained from the chamber and covering devices are summarized as follows.[10,11]

- The deposition of SO_2 to a reactive surface, such as galvanized steel, is controlled by atmospheric resistance during periods of surface wetness. The deposition rate of NO_2 is an order of magnitude smaller than SO_2 and is likely controlled by surface re-

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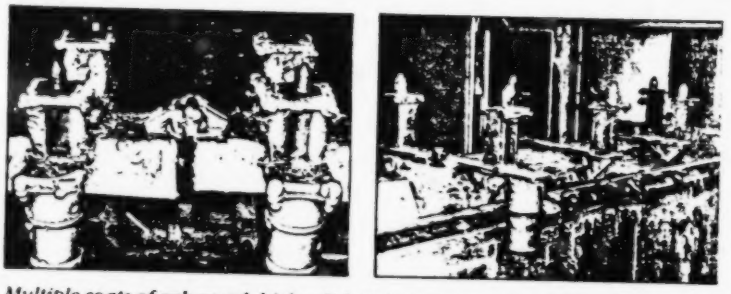

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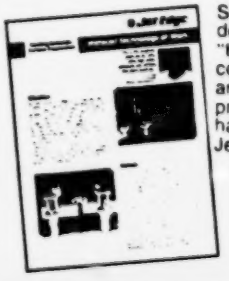

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sistance. To a dry surface SO_2 will likely deposit until a saturated monolayer is formed.

- Gaseous HNO_3 appears to deposit continuously to wet and dry surfaces.
- During periods of surface wetness, SO_2 and formaldehyde have similar rates of deposition to material surfaces. An adduct of formaldehyde and HSO_3^-, identified in Los Angeles smog, has been observed to have formed on material surfaces during chamber exposures.
- Chamber and field exposures indicated the deposition of SO_2 to a wet painted surface may be controlled by the level of oxidants such as H_2O_2.
- The dry deposit of SO_2 and HNO_3 dissolves alkaline components within paint film. Field exposures show that wet deposition (rain acidity) in rain also promotes dissolution of these components. $CaCO_3$, ZnO, and aluminum flakes appear to be susceptible to acidic deposition.
- Analysis of runoff collections from the field covering devices indicates that the atmospheric deposition of basic particles (e.g., $CaCO_3$) to materials may protect the material from acid deposition.

Analytical Techniques

Other researchers are working with EPA to develop the use of state-of-the-art analytical techniques by conducting short-term exposure experiments to determine whether acid deposition plays a role in film failure of coated substrates. Effects of acid deposition on coated steel substrates are being investigated at The Johns Hopkins University (JHU) and The Martin Marietta Laboratories (MML) in Baltimore, MD. An *in situ* electrochemical monitor based on electrochemical impedance spectroscopy (EIS) is being developed for detection of film degradation during atmospheric exposure. The investigation of the effects on coated wood substrates is being conducted by The US Forest Product Laboratory (FPL) at Madison, WI and The North Carolina State University (NCSU) in Raleigh, NC. Fracture mechanics involving the use of uniform double cantilever beam (UDCB)

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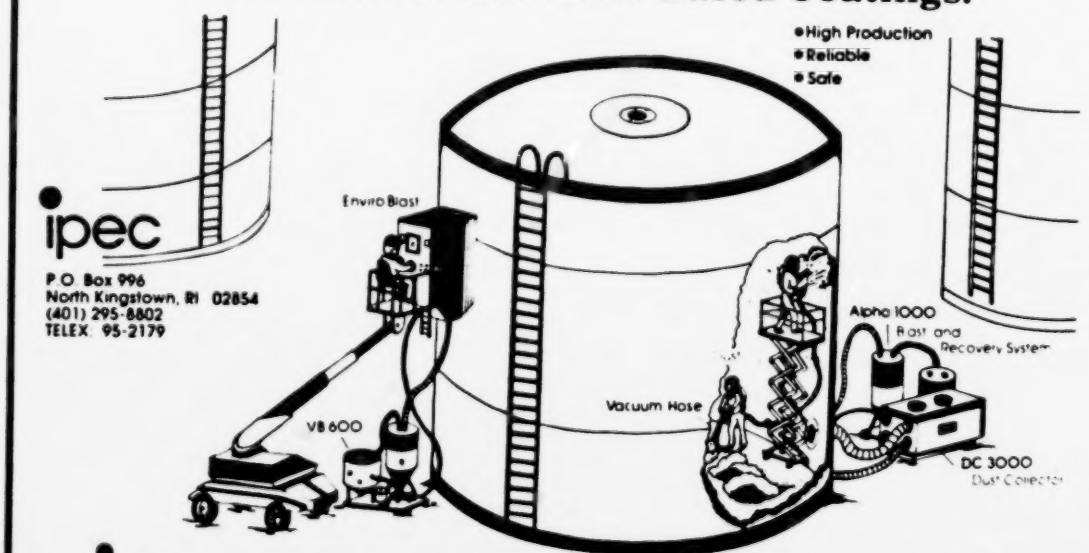

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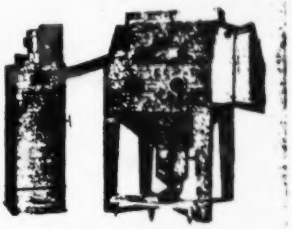

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specimens is being developed for testing the adhesion of painted wood specimens during exposure. In addition to the chamber facilities at EPA and NCSU, other facilities for exposing coatings to wet acidity and to gaseous pollutants have been developed at JHU, MML, and FPL. Analytical techniques and procedures that are under investigation by these researchers to determine early film substrate failures have previously been reported.[12,13,14,15,16] Major findings about the coated steel substrates that have been observed during short-term accelerated exposures within this phase of the coatings research program are summarized as follows.

- Before chamber exposure of alkyd steel specimens, cohesive failure is observed in the primer or topcoat at ambient conditions. Chamber exposures to humidity promote loss of adhesion between the paint primer and metal substrates. The addition of SO_2 accelerates the loss of adhesive strength of the specimens during chamber exposures.
- For alkyd steel specimens that were exposed to SO_2/condensation, brown spots formed within a few days after exposure. The discoloration occurred only on specimens that were mounted horizontally during the exposure. X-Ray Photoelectron Spectroscopy (XPS) analysis revealed sulfur to be present throughout the spot, while only trace amounts were present in areas around the spot.
- During sulfurous acid immersions, sulfur penetrated the topcoat and primer of the alkyd steel coating as well as the alkyd and latex coatings on wood substrates.
- Absorption and desorption experiments with SO_2 and the alkyd coating formulated for wood substrates revealed a significant amount of SO_2, about 10 percent to 20 percent of the amount absorbed, is retained by the film.
- In the presence of ultraviolet radiation (350 nm), SO_2 attacks the base terpolymer of the acrylic latex coating. This polymer was found to be stable upon exposure only to SO_2, while some degradation was observed with ultraviolet radiation.
- Weight loss measurements of the acrylic latex film with $CaCO_3$ indicate that all of the extender pigment is removed in 4 hours at pH 2.0 and in 430 hours at pH 4.0 sulfurous acid. Distilled water of pH 5.6 removed all the $CaCO_3$ in about 40 days. Removal of the $CaCO_3$ produced a reduction in the dynamic modulus of the film.

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During this phase of the coating exposure study, an array of analytical techniques as well as exposure facilities has been developed for characterizing the effects of acidic deposition on coated substrate systems. The next phase of study should involve using these techniques and exposure facilities to quantify the effect of ambient levels of acid deposition by conducting longer term exposures.

Conclusions

Within the NAPAP Materials Effects Program, unique chamber and field exposure facilities as well as an array of analytical techniques for environmental testing of coated substrate systems have been developed. With NAPAP ending in 1990, these unique facilities will be available for environmental testing and development of materials. By passing the Technology Transfer Act of 1986, Congress expressed a desire to make technology and facilities developed by the government available to private industry and other organizations through cooperative research and development agreements. By this mechanism, industry can fund government agencies to use facilities that would not otherwise be available. It is believed that the facilities and test procedures that have been developed within the NAPAP Materials Effects Program offer a unique capability for environment testing and development of materials of interest to private industry as well as other government agencies. Considerable information about environmental effects on coatings as well as other materials can be acquired during relatively short exposure periods.

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